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| 学位授与の題目 | Search for an Isomer State of $^{229}$Th with Extremely Low Energy using Alpha-Spectrometry<br>（α線スペクトロメトリーを用いた$^{229}$Thの極低エネルギー核異性体の探索） |
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## 学位論文要約

The author employed a rapid preparation method of α-source with high-resolution, and searched for $^{229m}$Th, a nuclear isomer with ultra-low excitation energy, produced from β-decay of $^{229}$Ac and α-decay of $^{233}$U. At first, the author developed a co-precipitation method which is by no means inferior to electrodeposition method in point of resolution (< 20 keV), chemical yield (~95 %) and uniformity. The greatest improvement over the other methods is that the newly devised method makes it possible to prepare an α-source within 5 minutes. Subsequently, the author produced $^{229m}$Th in the nuclear reaction $^{232}$Th(γ,p2n)$^{229}$Ac, followed by disintegration to $^{229m,g}$Th. The isomer was searched for by α-spectrometry. As a result, the half-life of $^{229m}$Th was estimated not to be longer than several days. However, a good counting statistics is not achieved by using the production of $^{229m}$Th in nuclear reaction. In addition, the author produced $^{229m}$Th by α-decay of 93mg of $^{233}$U and attempted to estimate its half-life. Finally, the author has concluded that the half-life of $^{229m}$Th is shorter than 1 hour at 1σ confidence level and shorter than 3 hours at 2σ confidence level, suggesting that an electron bridge mechanism or a medium effect is correlated with it.

## 学位論文概要

The purpose of this study is detection of the decay signal from an excited state of $^{229}$Th ($^{229m}$Th) with the energy lower than for any known nuclide. The excitation energy was estimated about 3.5 eV, which is extremely low with around $10^{-2}$～$10^{-6}$ of general atomic nucleus. Internal conversion process is prohibited in $^{229m}$Th because the excitation energy of the isomeric transition from $^{229m}$Th to $^{229g}$Th is lower than even binding energy of the outer-shell electrons in a thorium atom, the first ionization potential of which is 5.9 eV. Therefore, the investigation of the $^{229m}$Th decay is expected to provide a strong verification of an electron-bridge mechanism (EBM). In addition, since the probability of the EBM depends on the energy state of the outer-shell electrons, the half-life of $^{229m}$Th can vary with its chemical state; hence the isomer is also interesting from a chemical viewpoint. However, it is believed that no one has thus far succeeded in detecting a decay signal from $^{229m}$Th.

In this study, the author focused attention on α-decay as a direct decay signal from $^{229m}$Th. The expected favored α-decays from $^{229m}$Th had energies higher than those of $^{229g}$Th. This implies that the partial half-life of the α-decay of $^{229m}$Th is shorter than that of $^{229g}$Th. A significant finding is that α-particles from $^{229m}$Th are roughly distinguishable from those from $^{229g}$Th, if the α-spectrometry could be carried out with a good resolution. Therefore, the author (1) developed a rapid preparation method of α-source with high-resolution, and then (2) searched for $^{229m}$Th produced from β-decay of $^{229}$Ac and α-decay of $^{233}$U.

**(1) Development of a rapid preparation method of α-source with high-resolution**

In this study, the author developed a co-precipitation method which is by no means inferior to electrodeposition method in point of resolution (< 20 keV), chemical yield (~95 %) and uniformity. The greatest improvement over other methods is that the method is able to prepare an α-source within 5 minutes, even if the solution containing α-activity is about 20 mL. Therefore, the method is suitable to measure short-lived actinide elements.

**(2) Search for $^{229m}$Th produced from β-decay of $^{229}$Ac and α-decay of $^{233}$U**

The author produced $^{229}$Th in the nuclear reaction $^{232}$Th(γ,p2n)$^{229}$Ac, followed by disintegration to $^{229m,g}$Th. The $^{229m}$Th α-decay was searched for. As the results, the half-life of $^{229m}$Th was estimated that it was not long half-life but less than several days. However, a sufficient quantity of counting statistics is not able to be obtained by using production of $^{229m}$Th in nuclear reaction. In consequence, more restricted half-life can not be obtained in this way. Hence, the author attempts to estimate that of $^{229m}$Th by means of the α-ray spectrometry of $^{229m}$Th produced by the α-decay of $^{233}$U. Being produced from several milligrams of $^{233}$U, a large quantity of $^{229}$Th can be produced than using the nuclear reaction.

The author produced $^{229m}$Th by α-decay of 93mg of $^{233}$U and attempted to estimate its half-life. However, α-events were not obtained in the expected energy region for $^{229m}$Th, with the exception of those derived from $^{229g}$Th. From this experiment, the author has concluded that the half-life of $^{229m}$Th is shorter than 1 hour at 1σ confidence level and shorter than 3 hours at 2σ confidence level. In the framework of single-particle transition model, the partial half-life of direct photon emission from $^{229m}$Th is from 55 minutes to 16 hours in the energy range from 2.5 eV to 6.5 eV. If $^{229m}$Th is disintegrated only through the direct photon emission, the excitation energy of $^{229m}$Th should be more than 4.5 eV, corresponding to the half-life of 3 hours. In other words, the excitation energy of $^{229m}$Th is less than the energy, $^{229m}$Th disintegrated through not only the direct photon emission but also other decay channels, for example an EBM and a medium effect. A larger starting amount of $^{233}$U and many more experiments would provide more detailed half-life of $^{229m}$Th. In addition, to investigate the decay properties of $^{229m}$Th, it is important that the detailed excitation energy of $^{229m}$Th is investigated by photon detection and so on.

# 学位論文審査結果の要旨

本研究の対象である核異性体 $^{229m}$Th は原子核の励起エネルギーが約 3.5eV と推定されており、一般的な原子核の準安定状態に比べ極端に低い励起エネルギーをもつ。このエネルギーはトリウム原子の第一イオン化エネルギー（5.9eV）より低いために内部転換過程が禁止され、予言のみで実証がされていない「電子架橋過程」の非常に良い実証の場として期待される。この過程が興味深いのは、化学状態により核壊変の半減期が変化する可能性である。しかし、現在までに複数の研究グループによって試みられた $^{229m}$Th の核異性体遷移に伴う光子の検出は成功していない。このような状況で本研究の特徴は $^{229m}$Th の壊変信号として α 壊変に着目した点である。$^{229m}$Th からの α 壊変のエネルギーは妨害となる $^{229g}$Th のそれに比べて大きく、半減期も短いので微弱な $^{229m}$Th の α 線を分解能良く測定する技術があれば、その検出が可能であると考えられる。そこで論文提出者は自ら高いエネルギー分解能で α 線を測定するための線源調製法を開発し、$^{232}$Th（γ、p2n）反応による $^{229}$Ac の β－壊変や $^{233}$U の α 壊変で製造した $^{229m}$Th の探索を行なった。その結果、論文提出者は明瞭な事象を確認することはできなかったが水酸化物の化学状態にある $^{229m}$Th の半減期を 1 時間以下（信頼限界１σ）または３時間以下（同２σ）と求めることに成功した。これらの上限値は、γ 遷移の理論的予測と比較して短い値を与え $^{229m}$Th の壊変に電子架橋過程が寄与していることを示した。以上の成果を審査した結果、論文提出者は博士（理学）の学位に値すると判定したので、ここに報告する次第である。